Relating localized nanoparticle resonances to an associated antenna problem

Shakeeb Bin Hasan,[1] Carsten Rockstuhl,[1] Ralf Vogelgesang,[2] and Falk Lederer[1]

[1] *Institute for Condensed Matter Theory and Solid State Optics, Friedrich-Schiller-Universität Jena Max-Wien-Platz 1, 07743, Jena, Germany*

[2] *Max Planck Institute for Solid State Research Heisenbergstr. 1, 70569, Stuttgart, Germany*

We conceptually unify the description of resonances existing at metallic nanoparticles and optical nanowire antennas. To this end the nanoantenna is treated as a Fabry-Perot resonator with arbitrary semi-nanoparticles forming the terminations. We show that the frequencies of the quasi-static dipolar resonances of these nanoparticles coincide with the frequency where the phase of the complex reflection coefficient of the fundamental propagating plasmon polariton mode at the wire termination amounts to π. The lowest order Fabry-Perot resonance of the optical wire antenna occurs therefore even for a negligible wire length. This approach can be used either to easily calculate resonance frequencies for arbitrarily shaped nanoparticles or for tuning the resonance of nanoantennas by varying their termination.

PACS numbers: 41.20.Cv, 42.25.Fx, 73.22.Lp, 78.20.Bh

Small particles are among the earliest cases tackled by light scattering theory. The quasi-analytical rigorous solution for spheres dates back to the pioneering work of Gustav Mie in 1908 [1]. This approach can be considerably simplified if the size of the spheres is small compared to the illuminating wavelength resulting in the quasi-static approximation [2, 3]. The resulting analytic formulae for the polarizability of the sphere exhibit resonant denominators, such as the well-known expression $\varepsilon_\mathrm{m}(\nu) + 2\varepsilon_\mathrm{d}(\nu) = 0$ for the dipole resonance of a metallic sphere in a dielectric host medium. In this case and generally, the permittivities of the spheres and their surroundings need to exhibit opposite signs at resonance. The scattered field exhibits strongly enhanced, stationary evanescent components at the interface – a phenomenon which is termed localized surface plasmon polariton (LSPP) [4]. Intriguingly, it was shown that in this approximation the quality factor of the resonance solely depends on the material properties rather than the particle shape [5] which, however, affects the resonance frequency. Only for a few other particle shapes, e.g. ellipsoids and spherical shells or particles with a lower dimensionality, i.e. a cylinder, the resonance condition can be put in a similar form known from the sphere. The exploitation of these LSPPs at nanoparticles of different shape has led to various applications and is forming one branch of the prospering field of plasmonics.

If the metal is a perfect conductor, another resonance is supported by metallic wires if their length corresponds to a multiple of half the illumination wavelength. Such metallic wires constitute the basic building blocks of radio-frequency (RF) antennas. Recently, their downscaling into the visible attracted considerable interest and the field of optical antennas is now similarly established. Conceptually, optical antennas differ from RF antennas in that the field propagating along the wire is no more purely photonic but forms another polaritonic excitation [6]. This type of quasiparticle is referred to as propagating surface plasmon polariton (PSPP) due to its sole energy transport into propagation direction. As for any guided mode phenomenon, the PSPP dispersion relation may strongly depend on the wire's cross-section (see e.g. [7]).

The origin of resonances in these finite-length nanowires is well-understood in terms of Fabry-Perot resonances of the PSPP mode confined between the partially reflecting wire terminations [8–12]. Unlike in antennas at microwave frequencies, here the reflection coefficients are complex-valued, providing an additional phase term which mimics an increase of the wire length and depends on the actual shape of the termination. This resembles the situation in a planar Fabry-Perot resonator with Bragg mirrors, where the number of layers also affects the actual phase shift and thus the resonance condition. This is also the reason why multiples of half the resonance wavelength differ from the wire length [8, 10–13]. This peculiarity evoked research interest and both analytical and numerical results on the spectral and geometrical dependence of reflection coefficients were reported for abrupt or flat nanowire terminations [14, 15]. Moreover, associated geometries, such as e.g. trenches, grooves or slits on or in flat metal surfaces or metallic thin films, were analyzed with respect to their reflection/transmission properties of PSPP launched along the metal surfaces [16]. It allows obtaining insights into the underlying physics of phenomena observable in such systems. Examples thereof would be, e.g., the enhanced transmission in subwavelength apertures [17].

In short, to date metallic nanoparticles are largely studied in terms of LSPP resonances whereas wire nanoantennas are commonly analyzed in terms of PSPP standing wave phenomena and these seemingly disparate approaches have not been systematically integrated. A few reports on variable length nanoantennas [13, 18] con-

sider spherical particles as the limiting case of cylindrical wires. Nevertheless, it is challenging to disclose the underlying mechanism behind this convergence of resonances in general geometries at a physical level and to potentially exploit it for more complex nanoparticle geometries. Here we attempt to provide a unifying view. It will turn out that the LSPP resonances of arbitrary nanoparticles follow straightforwardly from solving the reflection problem at the nanowire termination.

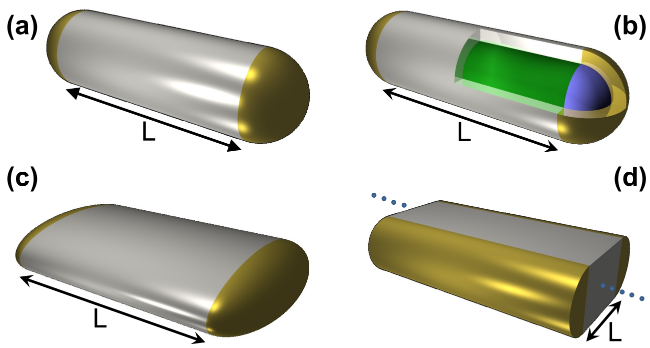

FIG. 1: Sketch of considered metallic nanoantenna geometries. Circular cylinder (a) and cylindrical shell (b) with a hemispherical termination; elliptical cylinder with a semi-elliptical termination (c); one-dimensional nanoantenna with a semi-cylindrical termination (d). All nanoentannas are composed of a nanowire (metallic color) and a termination (golden) while L is the nanowire's length

In the following we will consider various geometries to prove the universal nature of our considerations, but without loss of generality we are beginning with the circular nanowire with hemispherical terminations at both ends [Fig. 1(a)]. This nanoantenna becomes a sphere for vanishing wire length L. We hypothesize that the dipolar resonance of a sphere is caused by the constructive interference of the forward and backward propagating fundamental PSPP modes ($m = 0$) of the nanowire[19–21]. Then, the necessary condition for a Fabry-Perot resonance – that is, the round trip reproduction of the phase factor – is a total phase accumulation of an integer multiple of 2π. This condition can already be met for a wire with negligible length, i.e., the sphere, and requires a phase shift of π upon reflection at both terminations. In general, higher order PSPP modes do not need to be considered since all modes with $|m| \geq 2$ cut-off below a threshold wire radius [22]. Moreover, the PSPP mode with $|m| = 1$ diverges for a vanishing radius, thereby suffering from increasing radiation losses. Therefore, it cannot be excited anymore [23].

Thus, to generalize the idea, our claim is that it suffices to calculate the reflection problem of a PSPP at the respective termination and to look for a π phase shift to identify the LSPP resonance for the respective nanoparticle shape. Although detailed in the following only for the dipolar resonance, higher order resonances for particles with a non-spherical shape can be understood either as higher order Fabry-Perot resonances of the lowest order PSPP mode or lowest order Fabry-Perot resonances of higher order PSPP modes.

We used Comsol Multiphysics to numerically solve the reflection problem at the wire termination. The metallic nanowire is assumed to be semi-infinite and it is surrounded by a dielectric medium with a permittivity ε_{d}. The computational cell is enclosed by perfectly matched layers to mimic an open space. Silver (Ag) was used as the metal and its dispersive permittivity was fully considered [24]. The cylindrical wire had a radius of 10 nm. The exact value of the radius is not important provided that it is much less than the wavelength.

First we calculate the dispersion relation of the fundamental wire eigenmode and subsequently use this mode as illumination of the termination of the semi-infinite wire. Then the total (incident and scattered) field is calculated in a plane normal to the wire axis and located at $z = 0$. The complex reflection coefficient of this mode at the termination is extracted by using the azimuthal magnetic field $H_{\phi,\mathrm{tot}}(\rho, z)$ and by evaluating the overlap integral

$$r = -\exp^{-i2\beta l} \frac{\int_0^\infty H^\star_{\phi,0}(\rho,0)\left[H_{\phi,\mathrm{tot}}(\rho,0) - H_{\phi,0}(\rho,0)\right]\rho\,\mathrm{d}\rho}{\int_0^\infty H^\star_{\phi,0}(\rho,0) H_{\phi,0}(\rho,0)\rho\,\mathrm{d}\rho}, \tag{1}$$

where $H_{\phi,0}(\rho, 0)$ is the ϕ component of the incident PSPP mode ($m = 0$) at $z = 0$, β is the associated propagation constant and l is the distance between origin ($z = 0$) and termination. All quantities, except the wire geometry, depend on frequency. In passing we note that the distinction of what belongs to the wire and what belongs to the termination is arbitrary to a certain extent. The phase accumulated due to propagation and the phase accumulation due to reflection can easily be merged. However, since we wish to discuss solely the properties of the termination, the length $l = L$ is understood as the length of the nanoantenna along which no change of the cross-sectional profile occurs.

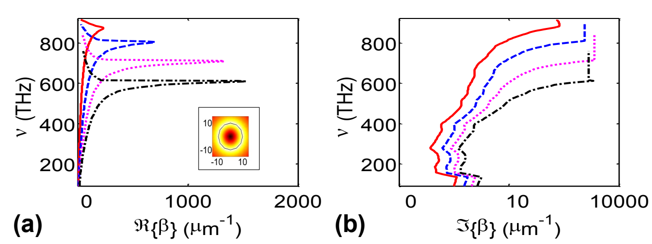

FIG. 2: Real (a) and imaginary part (b) of the propagation constant β of the lowest order PSPP mode as a function of frequency ν for selected values of ε_{d}; $\varepsilon_{\mathrm{d}} = 1$ (solid red), $\varepsilon_{\mathrm{d}} = 2.8$ (dashed blue), $\varepsilon_{\mathrm{d}} = 5.8$ (dotted magenta), $\varepsilon_{\mathrm{d}} = 9$(dotted-dashed blue). The inset shows the H_ϕ-field norm for a core radius of 10nm and $\varepsilon_{\mathrm{d}} = 1$.

In Fig. 2 the complex-valued propagation constant β of the lowest order PSPP mode is displayed as a function of the frequency and the permittivity of the surrounding medium. The real part exhibits the usual dispersion characteristic where the propagation constant increases with frequency until back-bending sets in. This back-bending is associated with a strongly increasing damping (imaginary part). In the succeeding spectral domain, any analysis of the reflection coefficient tends to be cumbersome since dissipation will entirely dominate the system.

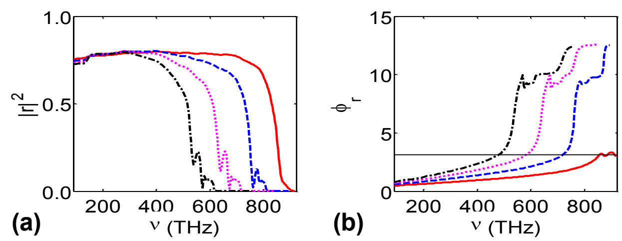

FIG. 3: Amplitude (a) and phase (b) of the reflection coefficient of the nanowire for selected values of ε_d; $\varepsilon_d = 1$ (solid red), $\varepsilon_d = 2.8$ (dashed blue), $\varepsilon_d = 5.8$ (dotted magenta), $\varepsilon_d = 9$ (dotted-dashed blue). The horizontal black line in (b) serves as a guide to eye and indicates where the phase of the reflected amplitude corresponds to π.

Figure 3 shows the complex reflection coefficient as a function of frequency for different ε_d extracted from the simulation of a semi-infinite wire for the respective lowest order PSPP mode. It can be seen that at low frequencies the modulus is constant and large with a phase shift around zero, suggesting a perfect metal-like behavior. The phase increases with frequency and undergoes an abrupt change at a critical frequency. This jump is associated with the decrease in the reflected amplitude and it appears in the frequency interval where back bending occurs. Now it is easy to extract the frequency where the phase jump of π occurs and to compare it to the resonance frequency predicted by the quasi-static theory for a small sphere. Additionally, it can be seen that the phase surpasses even values corresponding to multiples of π. Such frequencies would be associated to higher-order resonances as sustained by the nanoparticle with an even higher quality factor. Usually, such resonances are not observed, because they are dipole forbidden and exhibit excessive damping.

Figure 4(a) shows a comparison of the resonance frequencies predicted by the quasi-static theory as well as the PSPP reflection calculation for different surrounding media. The excellent agreement between both approaches demonstrates that the resonances of a sphere in the quasi-static limit are directly related to the corresponding limit of the nanoantenna problem.

To further investigate the universality of this conclusion, we briefly analyze other particle geometries in the following. Another special case of a spherical nanoparticle is one with a dielectric core and a metallic shell. It is of enormous practical relevance since its resonance frequency can be tuned across the entire visible spectral region by varying the metallic shell thickness [25]. These particles exhibit lower and higher energy LSPP resonances which appear as a result of the hybridization of individual resonances of the constituting metallic sphere and dielectric void [26]. We approximate the structure similarly as before by a nanowire made of a dielectric core surrounded by a metallic shell terminated with a hemispherical shell of the same construction [Fig. 1(b)]. Intuitively, we guess that the lower (upper) branch of the fundamental PSPP mode of the cylindrical metallic shell wire [27] is responsible for the lower (higher) resonance frequency of these particles. We repeat the aforementioned analysis for the lower branch PSPP mode and show a similar comparison between resonance frequencies predicted by quasi-static approximation and the reflected field where $\phi_r = \pi$ in Fig. 4(b) as a function of the ratio b/a between shell and core radii, respectively. An analysis of the highly dissipative upper branch was not attempted due to the conceptual difficulties associated with overdamped PSPPs (see also Fig. 2).

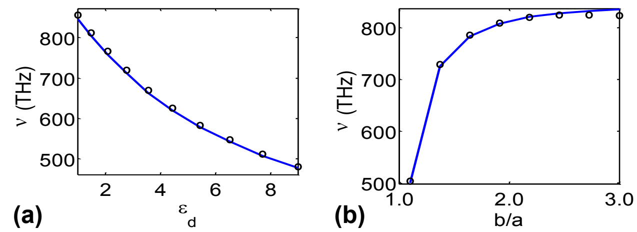

FIG. 4: Resonance frequency of a sphere (a) with a radius of 10nm as a function of permittivity of surrounding medium and of a spherical core shell particle (b) as a function of the ratio between shell and core radii b/a. The core radius was kept constant at $a = 10$nm. The permittivity of both the core and the surrounding medium was set to $\varepsilon_d = 1$. Dots correspond to resonance frequencies as extracted from the phase of the reflection coefficients and the solid lines correspond to the predictions from quasi-static theory.

Having substantiated our principle for the case of spherical particles associated with quasi-one-dimensional antennas, we now extend the scope of our study toward non-spherical geometries. First we consider an elliptical geometry (see Fig. 1(c)). They can be envisioned straightforwardly to be composed of an elliptical nanowire (radii a and b) of negligible length and terminations consisting of rotational symmetric semi-ellipsoids [Fig. 1(c)]. Note that the resonance frequencies of ellipsoids depend upon the illuminating polarization[28]. We set the semi-axis a to be the symmetry axis and present the results for the electric field polarized both perpendicular and parallel to it. Figure 5(a) shows the resonance frequency as calculated from the phase of the reflected

field at the wire termination compared to the predictions from quasi-static theory. In the antenna simulation the polarization is chosen by selecting the eigenmode for the respective illumination. For the illumination perpendicular to the semi-axis a (solid blue curve), we can recognize minor deviations for small values of b/a. This can be attributed to the fact that the quasi-static approximation is becoming worse for larger a. However, overall we see again an excellent agreement in the predicted resonance frequencies by both methods.

Lastly, Fig. 5(b) displays the comparison of the results obtained for a one-dimensional nanoantenna with semi-cylindrical terminations (see Fig. 1(d)) illuminated normal to the cylinder axis ($k_{||} = 0$) which is related to quasi-two-dimensional antennas. As can be seen from Fig. 1(d), the associated antenna consists of an insulator-metal-insulator (IMI) waveguide infinitely extended into one dimension. The thickness of the nanowire is 20 nm. To form the nanoantenna, the waveguide is terminated by semi-circular infinite cylinders [see Fig. 1(d)]. The IMI strip waveguide is well-known to support hybridized symmetric and anti-symmetric PSPPs [4], with the latter being strongly delocalized in the limit of a vanishing thickness (long-range surface plasmon polariton). The symmetric mode, on the contrary, localizes increasingly with decreasing thickness thereby standing out as the plausible source of LSPP resonance of cylinders in the quasi-static limit.

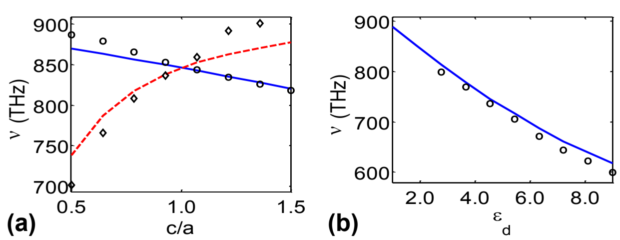

FIG. 5: Resonance frequency in (a) of an ellipsoid with $b = c = 20$nm surrounded by $\varepsilon_d = 1$ as a function of the ratio c/a between semi-axes and in (b) of a cylinder with a radius of 10nm as a function of ε_d. In (a), solid blue (dashed red) curve corresponds to quasi-static resonance of the spheroid under illuminating polarization perpendicular (parallel) to semi-axis a. Circular (diamond) marks indicate frequencies at $\phi_r = \pi$ for polarization perpendicular (parallel) to semi-axis a. The polarization in the antenna simulation is selected by choosing a respective wire mode as the illuminating field. In (b), solid blue curve corresponds to quasi-static resonance while circular marks denote the frequencies at which $\phi_r = \pi$.

Figure 5(b) shows the comparison between resonance frequencies predicted by the quasi-static limit ($\varepsilon_\mathrm{m}(\nu) + \varepsilon_\mathrm{d}(\nu) = 0$) and the frequencies where $\phi_r = \pi$ for various values of the permittivity of the surrounding dielectric. Simulations were done by using the symmetric mode for the illumination. Again, excellent agreement can be seen between the two approaches. We note here that we could not observe any $\pi-$crossing in phase ϕ_r of reflection coefficient for $\varepsilon_d = 1$ and 1.9. The reason is that the resonance frequencies occur in a spectral domain near the bulk plasma frequency where strongly increasing absorptive losses deteriorate the analysis of a reflection coefficient. The low propagation length of the surface plasmon polariton does not allow a reliable analysis,which is, after all, an intrinsic problem to such plasmonic systems. The increasing permittivity of the surrounding, however, causes a red shift of the resonance where dissipation is reduced.

In conclusion, we have proposed a novel perspective to look at the LSPP resonances of arbitrary metallic nanoparticles and relate them to resonances of optical nanoantennas. We show that LSPP resonances appear at frequencies where the phase jump upon reflection of PSPPs amounts to π. Numerical studies show that the resonance frequencies coincide with those obtained for LSPPs in the quasi-static approximation provided that this approximation is valid. Whilst shedding some new light on the relation between optical antennas based on nanoparticles and nanowires, our method establishes a systematic approach to manipulate the resonance position of optical antennas by suitably modifying the wire termination. This can be alternatively understood as a new degree of freedom, in addition to the wire cross-section, in the design of nanoantennas, which affects the propagation constant of PSPPs. For example, it is straight forward to imagine to equip, in perspective, nanoantennas with two different wire terminations and to obtain a further degree of freedom to adjust the resonance position of the nanoantenna. Moreover, we foresee that further research devoted to the question how to relate the reflection phase to measurable scattering quantities will be a fruitful direction. It can be anticipated that this approach may both promote and unify the fields of localized plasmon polaritons and optical nanoantennas.

Financial support by the German Federal Ministry of Education and Research (PhoNa), by the Thuringian State Government (MeMa) and the German Science Foundation (SPP 1391 Ultrafast Nano-optics) is acknowledged. We would like to thank S. Fahr for assistance in preparing some figures.

[1] G. Mie, Annalen Der Physik **25**, 377 (1908).
[2] C. F. Bohren and D. R. Huffman, *Absorption and scattering of light by small particles* (Wiley-Interscience, New York, 1983).
[3] D. R. Fredkin and I. D. Mayergoyz, Physical Review Letters **91**, 4 (2003).
[4] S. A. Maier, *Plasmonics: Fundamentals and Applications* (Springer, New York, 2007).
[5] F. Wang and Y. R. Shen, Physical Review Letters **97**, 4 (2006).

[6] J. J. Hopfield, Physical Review **112**, 1555 (1958).
[7] E. Feigenbaum and M. Orenstein, Optics Express **14**, 8779 (2006).
[8] L. Novotny, Physical Review Letters **98**, 4 (2007).
[9] E. S. Barnard, J. S. White, A. Chandran, and M. L. Brongersma, Optics Express **16**, 16529 (2008).
[10] T. Søndergaard, J. Beermann, A. Boltasseva, and S. I. Bozhevolnyi, Physical Review B **77** (2008).
[11] J. Dorfmüller, R. Vogelgesang, R. T. Weitz, C. Rockstuhl, C. Etrich, T. Pertsch, F. Lederer, and K. Kern, Nano Letters **9**, 2372 (2009).
[12] J. Dorfmüller, R. Vogelgesang, W. Khunsin, C. Rockstuhl, C. Etrich, and K. Kern, Nano Letters **10** (2010).
[13] G. W. Bryant, F. J. G. De Abajo, and J. Aizpurua, Nano Letters **8**, 631 (2008).
[14] S. J. Al-Bader and H. A. Jamid, Physical Review B **76**, 7 (2007).
[15] R. Gordon, Optics Express **17**, 18621 (2009).
[16] L. H. Lalanne P., Hugonin J.P. and W. B., Surface Science Reports **64**, 453 (2009).
[17] H. T. Liu and L. P., Nature **452**, 728 (2008).
[18] J. Aizpurua, G. W. Bryant, L. J. Richter, F. J. G. de Abajo, B. K. Kelley, and T. Mallouk, Physical Review B **71**, 13 (2005).
[19] J. C. Ashley and L. C. Emerson, Surface Science **41**, 615 (1974).
[20] C. A. Pfeiffer, E. N. Economou, and K. L. Ngai, Physical Review B **10**, 3038 (1974).
[21] H. Khosravi, D. R. Tilley, and R. Loudon, Journal of the Optical Society of America a-Optics Image Science and Vision **8**, 112 (1991).
[22] D. E. Chang, A. S. Sorensen, P. R. Hemmer, and M. D. Lukin, Physical Review B **76**, 26 (2007).
[23] Z. P. Li, K. Bao, Y. R. Fang, Y. Z. Huang, P. Nordlander, and H. X. Xu, Nano Letters **10**, 1831 (2010).
[24] P. B. Johnson and R. W. Christy, Physical Review B **6**, 4370 (1972).
[25] S. J. Oldenburg, R. D. Averitt, S. L. Westcott, and N. J. Halas, Chemical Physics Letters **288**, 243 (1998).
[26] E. Prodan, C. Radloff, N. J. Halas, and P. Nordlander, Science **302**, 419 (2003).
[27] S. J. Al-Bader and M. Imtaar, Journal of the Optical Society of America B-Optical Physics **10**, 83 (1993).
[28] T. Okamoto, in *Near-Field Optics and Surface Plasmon Polaritons* (Springer, Berlin-Heidelberg, 2001).